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ToWhat Extent Can Maillard Reaction Products Influence the Probiotic and Harmful Bacteria?

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MAILLARD reactions (MRs) is a non-enzymatic browning process, which helps to produce flavor and aroma, and different compounds with different health effects in the food and feed. The reaction is between reducing carbohydrates (reducing sugars) and amino compounds and it can be classified into three stages (initial, intermediate, and final). Depending on the reaction and storage conditions, activity and complexity of reactants used tools for environment control, and applied biotechnological applications, both beneficial and toxic MRPs can be produced. This review aims to analyze the activities and mechanisms of components forming by the Maillard reaction and the most common bioactive Maillard reaction products (MRPs) with prebiotic, antifungal, and antibacterial activity. Moreover, the study of the possible formation of toxic chemicals and having a look at the elimination ways of this potential problem was also an important goal in this review. The main aim of this review is to address the benefits of Maillard reaction products, which have been identified in previous studies to have antimicrobial and prebiotic effects, for the treatment of human pathogens. The present study will help to understand the positive and negative sides of Maillard reaction.

Keywords: Melanoidin; Gut health; Amadori products; Antimicrobials; Prebiotics.

1. Introduction of the Maillard reaction

Maillard reactions (MRs) are very important and highly complex conjugations, which have already been applied in food science in several disciplines such as human pathology, physiology, flavour chemistry, and geology (Charnock et al. 2022). These reactions mainly deal with the transformation of both carbonyl and amine groups, via non-enzymatic condensation in a multi-step reaction cascade through consecutive and parallel sub-reactions (Liu et al. 2022b). Several studies have used MRs more than 100 years ago with a focus on different areas of research including protein processing in the food and pharmaceutical industry (Zhong et al. 2019; Nooshkam et al. 2020; Liu et al. 2022b).

The Maillard reaction (MR) is a non-enzymatic browning process, which is one of the most important chemical reactions in food processing and storage. It is a spontaneous and exceptional reaction between reducing carbohydrates (reducing sugars) and amino.

compounds, following the formation of intermediate compounds or poly-carbonyl compounds reacting with amino acids or proteins. MR is a highly complex interaction with the classification of three stages (initial, intermediate, final), first proposed by Hodge (Table 1). The Maillard reaction products (MRPs) have both beneficial and negative impacts on human health (Cha et al. 2019; Murata et al. 2021). The average human diet contains a significant amount of them (Nooshkam et al. 2019b).

Understanding the process of the glucosamine (N-substituted glycosylamine formation) rearrangements and the study of the reaction equations will help the

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understanding of the Maillard reaction. To study this highly complex reaction more experiments should be carried out justified within different model systems (between reducing sugars and amino acids) (Noshkham et al. 2019a; Liu et al. 2020; Murata 2021; Charnock et al. 2022; Lie et al. 2022).

TABLE 1. The chemical reactions and forming compounds during different stages of the Maillard reactions.

Stages of MRs	Maillard Reactions (MRs) and their forming compounds		
	Chemical reactions	Products	Color
Initial	Initial glycosilation (ribose-glycine condensation)	ARPs: Amadori products (intermediate of the AGEs)	Transparent
	Glycation (formed from alpha-oxoaldehydes	Advanced glycation end-products (AGEs)	Transparent
Intermediate	Sugar degradation, fragmentation, degradation of amino acids	Carbonyl compounds	Transparent/Yellow
Final	Heyn's rearrangement products from ketose sugars condensation of aldols, condensation of aldehyde-amine, formation of heterocyclic nitrogen compounds	Melanoidins	Brown

(Charnock 2022; Shahidi and Hossain 2022; Singh et al. 2021a; Cui et al. 2021)

2. Forming compounds by Maillard reactions

2.1 Maillard reaction and its stages

The MR results in the initial stages of Schiff base formation and rearrangement to amadori (1-amino-1-deoxy-2-ketose from aldose sugars, ARPs) or Heyn'sproducts (2-amino-2-deoxyaldose from ketose sugars, HRPs) via 1,2-enaminol. The MRPs can form deoxyosones (α-dicarbonyl compounds), which react with additional nucleophiles and form Strecker aldehydes. ARPs considered to be a precursor of fragrant volatile compounds (Li et al. 2021) and browning precursors (Cui et al. 2021). MR forms advanced glycation end products (AGEs), which compounds have been implicated in kidney-related complications and have negative health effects, such as diabetes, oxidative stress, inflammatory processes, and alteration of proteins (Cha et al. 2019). In the first stage, the most reactive amino acid is lysine, which has higher importance, as it can degrade during the initial reactions due to heat treatment (Xiang et al. 2021).

At the intermediate stage of MR, the ARPs break down into different small pigments that highly depend on the pH. All of the routes are containing the formation of deoxyosones (reactive intermediate products), furthermore, furfural and reductones can be formed during this stage (Zhou and Langrish 2021). The highly reactive dicarbonyl compounds with yellow color and low molecular weight are forming at this step, when sugar degradation, fragmentation, and degradation of amino acids are befalling. The most common dicarbonyl compounds are glyoxal (GO), methylglyoxal (MGO), and diacetyl (DA) (Cha et al. 2019). Regarding other studies furfural (F) and hydroxymethylfurfural (HMF) (Rodier and Hartel2021), 5-methyl-2-furfural (MF), furylmethylcetone (FMC) can also form at pH 7 or lower (Liu et al. 2022b). The 5-Hydroxymethylfurfural (HMF), with simple molecular structures, is considered one of the most versatile platform molecules (Yang and Mu 2021; Zhai et al. 2021), which can be synthesized by the thermal decomposition of the sugars.

The products from the final stage brown pigments (melanoidin) with large polymeric compounds are formed by condensation of aldols, aldehyde-amine, and heterocyclic nitrogen compounds and contribute unique organoleptic properties (Nooshkam et al. 2019b; Murata 2021; Charnock 2022; Liu et al. 2022) In the late stage, during condensation, polymerization, degradation, and cyclization heterogenous brown pigment polymers can be formed (melanoidin) with high molecular weight (up to 100,000 g/mol) (Murata 2021). It can be said, that the exact reaction mechanisms and kinetics of the MR are unexplored, so further studies are necessary (Zhou and Langrish 2021).

2.2. Advantages of Maillard reaction

The amadori rearrangement products have importance in food science and application and have a great prospect of application as substitutes for monosodium glutamate (Cui et al. 2021). According to another study, during MR reductive ketones,

aldehydes, and heterocyclic compounds can be produced, which can be utilized as color and flavor in the food industry (Liu et al. 2020). The generation of aroma compounds is related to the MR, as products at different stages can act as a precursor and interact with other degradation constituents of food and produce several long-chain heterocyclic compounds during cooking (Shahidi and Hossain 2022).

Antioxidant products can be formed during MR, so the antioxidant activity can be improved (Nooshkam et al. 2019b).

The most important MRPs are the NεFructoselysine (FL) (early stage) and melanoidin (late stage). This reaction generates products that may have beneficial impacts **(Fig. 1)**, such as antioxidant, antimicrobial, prebiotic, and antihypertensive (Nooshkam et al. 2019b; Nasirpour and Saeidi 2021).

Some studies have shown that MRPs can inhibit the enzyme activity of fungi and have a similar anti-browning activity to that of metabisulfite. Moreover, fungi tyrosine has a partial and direct irreversible inhibition effect through chelating the copper ions in the active site of the enzyme successfully applied to improve the oxidative stability of diverse foods (Nooshkam et al. 2019b).

The 5-Hydroxymethylfurfural (HMF) forming during the intermediate stage has been widely used as a quality indicator during food processing and storage (Rada-Mendoza et al. 2022), furthermore, it is an indirect advantage, as it can be acceptable in some foods, due to its connection between organoleptic properties (Martins et al. 2022).

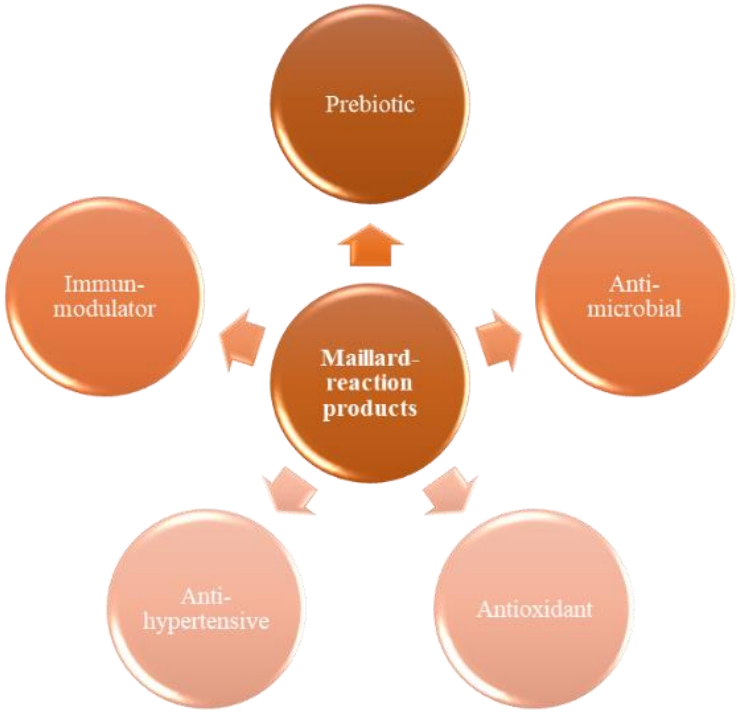

Fig. 1. Pharmacological effects of MRPs.

2.2 Problems of Maillard reaction

One of the main disadvantages is the alteration of the nutritional value of foods, like the reduction of the digestibility of proteins, the loss of lysine, the generating of mutagenic and toxic compounds (Nooshkam et al. 2019b) and may have negative health effects, like Alzheimer, cardiac disease obesity and diabetes (Nasirpour and Saeidy 2021). It has been reported, that cooking at high temperatures and time may increase the formation of undesirable chemicals (acrylamides, heterocyclic aromatic amines, polyaromatic hydrocarbons, advanced glycation end products) (Li et al. 2021b). According to some investigations by Food and Drug Administration (2021), a significant amount of potentially harmful chemicals (126 pieces) can be produced by the Maillard reaction (Jia et al. 2022).

Melanoidin has anionic (heavy metals) and complex maker (mutagenic, carcinogenic compounds) properties, so it can act as an environmental pollutant at high concentrations and is commonly produced by food, fermentation industries, and distilleries.

When melanoidin is released into the environment at high concentration they cause several threats to land and aquatic ecosystem, due to its recalcitrant coloring, anionic, offensive odour, dark color, and complex nature (Singh et al. 2021a).

Dark-color wastewater can be included very high total dissolved solids (TDS), phosphate, sulfate, phenolics, biological oxygen demand (BOD), chemical oxygen demand (COD), and heavy metals (Fe, Ni, Cu, Cr, Pb, Cd, Zn) (Sharma et al. 2021).

It is worth mentioning, that melanoidin is with other organic compounds in the distillery wastewater matrix, such as di-n-octylphthalate, di-butylphthalate, benzenepropanoic acid, and 2-hydroxycaproic acid, which compounds extend carcinogenic, mutagenic, genotoxic, and endocrine disruptors effects in the environment (Singh et al. 2021b).

The 5-Hydroxymethylfurfural (HMF), can cause different negative and positive health effects (Zhao et al., 2021).Some recent studies have demonstrated the toxic effects of the consumption of HMF, and pay attention to the importance of the dose intake in the case of human and animal health and the design of the strategy, health risk assessment, and more method development for to avoiding of negative sides (mutagenic, hepatotoxic, nephrotoxic, an inducer of neoplastic transformations, allergic, and cellular glutathione levels reducing effects) caused by HMF (Lee et al. 2019).

When MR is not controlled properly, advanced glycosylation end products (AGEs) can be formed, which can cause trigger inflammatory reactions, insulin resistance, and vascular damage, increase the level of oxidative stress, and active carbonyl stress (Jia et al. 2022), furthermore can play a role in some neurocognitive and mental disorders (D'cunha et al. 2022).

Maillard reaction can be influenced by selecting and maintaining proper reaction and storage conditions, activity and complexity of reactants, obtaining strict environment control, applying biotechnological applications, and developing new methods for the identification of toxic chemicals, as seen in **Fig. 2.** (Nooshkham et al. 2019b; Liu et al. 2020; Singh et al. 2021a; Murata 2021; Tripathi et al. 2021; Charnock et al. 2022; Li et al. 2022).

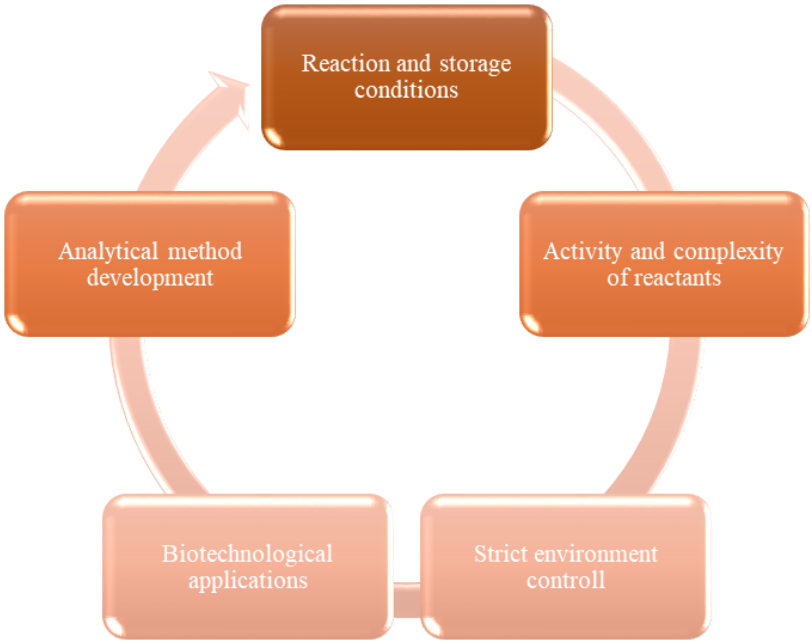

Fig. 2. Controlling of negative sides of the non-enzymatic browning process.

It has been argued that the composition of the final biobased large class of molecules depends on the reaction conditions (temperature, processing time, pH, water content, pressure, water activity), the activity, and the complexity of reactants (sulfites, amino compounds, and sugars).

Moreover, strict control is obligatory for the modeling of the quality and health properties of the final product and avoiding the possibility of forming toxic or carcinogenic chemicals, like furfural, furosine, and acrylamide (Nooshkham et al. 2019b;

Liu et al. 2020; Singh et al. 2021; Murata 2021; Charnock et al. 2022).

The storage on different relative humidity, temperature, and time can also affect the reactions (Li et al. 2021a) and should be properly planned and controlled.

Method development for the identification of HMFs and AGEs (Li et al. 2022) may also help to inhibit the levels of toxic MRPs. It is worth knowing that polyphenols are able to decrease acrylamide and 5-hydroxymethylfurfural (5-HMF) levels in the model system (Han et al. 2022). The usage of genistein (isoflavones, group of aglycones) pharmaceutical drugs can decrease the number of the forming AGEs (Sharfi-Rad et al. 2021).

Some microorganisms, for example, *Micrococcus, Flavobacterium, Pseudomonas, Bacillus, and Enterobacter* sp. have excellent biosorbent ability (high surface ratio, active chemisorption sites in their cell wall), which makes the great heavy metal tolerant and accumulators. Their usage can help reduce environmental toxicity in humans and plants (Tripathi et al. 2021).

3. Antimicrobial and prebiotic effects of Maillard reaction product

3.1 The importance of the MR in the health condition of the gut

Maillard reaction products have great nutritional and medicinal attributes, for example antioxidant, antimicrobial, prebiotic, and antihypertensive (Nooshkam et al. 2019b), however, the highest focus is on their valuable antioxidant potential (Habinshutiet al. 2019). Recently, the prebiotic activity of MRPs has become an important tool in the food industry due to its important role to improve and balance the composition of microbiota in the gut, which is linked to the health conditions of the host, especially intestinal health, immune modulation, pathogen invasion, and metabolic side effects (Pérez-Burillo et al. 2020; Zhong et al. 2021; Kukuminato et al. 2021). Furthermore, it is shown to have (**Table 2)** prebioticactivity by promoting the growth of probiotic bacteria (Kukuminato et al. 2021; Zhong et al. 2021; Pérez-Burillo et al. 2020).

TABLE 2. Antimicrobial and prebiotic effects of different MRP samples.

Activity	Name of the sample	Microorganisms tested	Reference
Antibacterial activities			
	Xylose-Phenylalanine-based melanoidin	*Bacillus cereus*	Kukuminato et al. (2021)
	Chitosan–inulin MRPs	*Bacillus subtilis*	Nooshkam et al. (2019a)
	Xylose-Phenylalanine-based melanoidin	*Brevibacillus brevis*	Kukuminato et al. (2021)
	Fermented Maillard reaction products made by milk proteins (FMRPs) (Probiotic bacteria: *Lactobacillus rhamnosus* and *Lactobacillus gasseri)*	*Clostridium perfringers*	Kim et al. (2021)
	Chitosan–inulin MRPs	*Escherichia coli*	Nooshkam et al. (2019a)
	D-xylose-l and Phenylalanine based melanoidin	*Listeria monocytogenes*	Kukuminato et al. (2021)
	Xylose-Prolin based melanoidin	*Listeria monocytogenes*	Kukuminato et al. (2021)
	Xylose-Prolin based melanoidin	*Salmonella* Typhimurium	Kukuminato et al. (2021)
	Proline-rich peptides in the melanoidin protein	*Staphylococcus aureus*	Kukuminato et al. (2021)
	MRPs-chitosan low pH value	*S. aureus*	Nooshkam et al. (2019a)
Antifungal activities			
	MRPs derived from xylose-cysteine-corn peptide hydrolysate (XCP)	*Pseudomonas aeruginosa*	Nooshkam et al. (2019a)
	MRPs-chitosan low pH value	*Aspergillus niger*	Nooshkam et al. (2019a)
	Chitosan–inulin MRPs	*Candida albicans*	Nooshkam et al. (2019a)
	MRPs-chitosan low pH value	*Aspergillus flavus*	Badr et al. (2022)
	Spent coffee grounds (75% of original coffee beans)	*Aspergillus ochraceus*	Badr et al. (2022)
Prebiotic activities			
	Spent coffee grounds (75% of original coffee beans)	*Bifidobacterium* spp.	Pérez-Burillo et al. (2020)
	Bread crust, pilsner and black beers, chocolate and sweet wine melanoidins	*Faecalibacterium* spp.	Pérez-Burillo et al. (2020)
	Biscuit melanoidins	*Lactobacillus brevis*	Pérez-Burillo et al. (2020)
	Digested glycoconjugates produced by the MR	*Lactobacillus casei*	Zhong et al. (2021)
	MR between Soy protein isolate and prebiotic oligosaccharides	*Lactobacillus* spp.	Kukuminato et al. (2021)

Antibiotic resistance has become an increasing number of patients with bacterial invasion placing critical health problems worldwide (Cars et al. 2021). Maillard reaction products can play a role in the fight against pathogenic invasion, as have antimicrobial inhibitory activity against a wide range of food-poisoning microorganisms.

The most significant prebiotic and antimicrobial agent from the group of MRPs is melanoidin (antioxidant) forming at the late stage (Nooshkam et al. 2020; Kukuminato et al. 2021; Shaheen et al. 2021), however, ε-polylysine (ε-PL) (Luz et al. 2019; He et al. 2021).

3.2. Antimicrobial and prebiotic compounds formed by MR

The high molecular weight (MW) brown macromolecular polymers (melanoidins) are a nitrogen and carbon source for the digestion of probiotics, a prebiotic agent that is selectively utilized by commensal bacteria, bringing about beneficial changes in the gut microbiota (Shaheen et al. 2021). Melanoidins have stronger antimicrobial properties against Gram-positive bacteria than Gram-negative, which is caused by the presence of an outer membrane. MRPs is shown to be very useful food preservative to control pathogenic bacteria (Nooshkam et al. 2020; Kukuminato et al., 2021). The structure of melanoidin is varied in different food (Pérez-Burillo et al. 2020) and the rate of browning can be increased by the raising temperature (Singh et al. 2021a).

The pentoses fasting of the MR due to the reaction time, in contrast to the hexoses with cyclic. The type of amino acid is also very important (Kukuminato et al. 2021). Melanoidins can contain different phenolic compounds (phytochemicals) and increase the absorption of phenolic compounds. The metabolization of these brown polymers by gut microbe consequences in the release of phenolic compounds, which can take out antioxidant properties (Pérez-Burillo et al. 2020) and have an antimicrobial effect (Panda and Duarte-Sierra, 2022).

The MRPs are oxygen scavengers, reactive oxygen scavengers, reducing agents, and metal chelating agents and thus can act as an antioxidant (Fotschki et al. 2022). The antimicrobial property of melanoidin especially due to their metal chelating features, antioxidant capacity, high surface activity, and inhibiting effect towards catabolic enzymes. Correlations found between the melanoidin content and antioxidant capacity (Martinez-Gomez et al. 2020).The growth of good bacteria affected by polyphenols. The strength of the effect dependson the type of the chemical (Fotschki et al. 2022).

Recently, there has been renewed interest in MRPs derived bioactive peptides (BPs) with functional antimicrobial and prebiotic effects, which depend on the size, sequence and amino acid composition, methods of production, molecular modification, and molecular interactions (Wu et al. 2021). The BPs are short amino acid sequences, that have physiological effects on the human body, and moreover recently utilized as functional food ingredients (Aursuwanna et al. 2022). The BPs have some problems, such as low antimicrobial activity and easy degradation by proteases, but MR is an excellent method to increase the functional properties (antimicrobial and antioxidant attitudes (Jiang et al. 2018).

The presence of peptide bonds within melanoidins and the incorporation into melanoidin polymers are suggested by some studies (Kukuminato et al. 2021).

Chitin can be formed into chitosan by MR through Schiff base linkage and Amadori or Heyns rearrangement.The quality and shelf life of food can be enhanced by MR. Furthermore, the antimicrobial, antioxidant, and emulsifying properties of chitosan can be strengthened by melanoidin products and reductones (Hafsa et al. 2021). The addition of glycated chitosan (Maillard conjugates) and inulin-chitosan conjugates to food products can help to produce food products with good microbiological status and long shelf-life (Nooshkam et al. 2019a; Nooshkam et al. 2020).

The ε-polylysine (ε-PL) is a small molecular weight, homo-polyamide compound with functional properties, like antimicrobial and antioxidant (He et al. 2021; Chie 2022), which could destroy cell walls and membranes of a broad spectrum of food pathogens and spoilage organisms (Luz et al. 2019; He et al. 2021). It is considered to be a safe food-grade antimicrobial agent and the ability depends on its cationic nature.

During electrostatic interactions, ε-PL supports cell membrane disruption (Luz et al. 2019). The ε-PL-dextran conjugates are successfully produced via Maillard reaction using a wet-heating method (90°C for up to 120 minutes).

According to Chie's (2022) investigation, 60 minutes period of heat treatment was shown to be optimistic for the conjugation, the preservation of their antimicrobial activity, and the improvement of their radical scavenging activity (Chie 2022). ε-PL is considered to be a green preservative, which is prolonging the shelf life of some seafood and is

produced by MR without solvents and catalysts, furthermore approved by the Food and Drug Administration (FDA).

Conclusion

The Maillard reaction products (MRPs) are widespread in the food and pharmaceutical industries. MRPs could bring imposing colors, and fragrances, and improve the taste of food. The MRPs are shown to be a successful tool to improve gut microbiota, avoid pathogen invasion, and have antibacterial resistance, as they have high antibacterial, antifungal, and prebiotic potential. The most important compounds, which play a role in this pharmacological effect are melanoidin, and ε-polylysine (ε-PL).

In the future, we are supposed to develop the applications of the MR of foods with bioactive peptides, chitin, and galactooligosaccharides, which are beneficial to us.On the other hand, there are now many methods to investigate the forming compounds by MR, but further method development is necessary to avoid unfavourable MRPs and explore new compounds, and study their reaction mechanisms.

Recently, MRPs with antimicrobial and prebiotic effects have just a little bit of attention, so further studies are necessary to obtain new scientific results in this area.

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